

IN THE CLAIMS

Please amend claim 2 as follows:

2. (Amended) The transparent laminate of claim 1, wherein a coating film of the self-emulsifiable emulsion (A) has an elongation of 240 to 500 % and a 100 % modulus of 190 kgf/cm² or more.

REMARKS

The foregoing amendments are submitted for the purpose of correcting errors which were made in the international application on which the present U.S. application is based. That is, these errors are not translation errors.

On page 5 of the specification, and in claim 2, the units for 100 % modulus have been changed to kgf/cm², which is consistent with the units for 100 % modulus disclosed at, for example, page 20, lines 14 and 30.

On page 7, line 2, "thallium oxide" has been changed to --tantalum oxide--. The oxides disclosed at the top of page 7 follow the symbols for the metals at page 6, lines 22-24, which include Ta (instead of Th).

The other change, at page 13, is self-explanatory.

Attached hereto is a marked-up version of the changes made to the Specification and claim 2 by the current amendment. The attached pages are captioned "Version with markings to show changes made."

Respectfully submitted,

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May 31, 2002

a composite oxide to be described hereinafter, the obtained film may be whitened or the impact resistance of an article coated with the coating film may lower. When a polyurethane having a bisphenol A skeleton is used in combination with composite oxide fine particles, the whitening of the film can be prevented and high impact resistance can be obtained.

The above polyurethane preferably has a number average molecular weight of 100,000 to 1,000,000.

The self-emulsifiable emulsion of the above polyurethane can be produced by adding the above polyurethane to an aqueous medium containing a basic compound such as an aliphatic amine or aromatic amine and stirring. The above polyurethane is emulsified to particles with diameter of 10 to 50 nm.

The solids content of the self-emulsifiable emulsion is adjusted to preferably 50 wt% or less, more preferably 20 to 48 wt%.

The self-emulsifiable emulsion used in the present invention may be acquired under the name of Neo Rez (trademark) of Abisia Co., Ltd. or Adecabontiter (trademark) of Asahi Denka Kogyo K.K.

The self-emulsifiable emulsion preferably gives a coating film having an elongation of 240 to 500 % and a 100 % modulus of 190 kgf/cm² or more.

Since the coating film becomes hard by the dispersion of inorganic oxide fine particles to be described hereinafter into the resin, the resin preferably has the above physical properties. A coating film having an elongation of more than 500 % has high thermoplasticity and the heat resistance of an article coated with this coating film tends to deteriorate. When the elongation is less than 240 %, the impact resistance tends to lower. When the 100 % modulus is 190 kgf/cm² or more, excellent impact resistance and heat resistance are obtained.

The component (B) used in the present invention is a

sol of an inorganic oxide having a hydrophobic group on the surface. The inorganic oxide fine particles of this sol increases the hardness, heat resistance and weatherability of the coating film. Inorganic oxide fine particles containing an oxide of Ti or Zr enhance the refractive index of the coating film and serves to prevent the generation of an interference fringe. The whitening of the coating film can be prevented by making a hydrophobic group existent on the surface of each inorganic oxide fine particle.

Examples of the hydrophobic group include an alkyl group, alkyl group substituted by an acryloxy group, alkyl group substituted by a methacryloxy group, alkyl group substituted by a glycidyloxy group and alkylene glycol group having an alkyl group at one terminal. Out of these, an alkyl group substituted by an acryloxy group and an alkyl group substituted by a methacryloxy group are particularly preferred because a coating film having high stability is obtained. The whitening of the coating film cannot be prevented when an amino group or an ureido group which is not a hydrophobic group is made existent on the surface of the inorganic oxide fine particle.

The inorganic oxide of the sol (B) is an oxide of at least one element selected from the group consisting of Si, Al, Sn, Sb, Ta, Ce, La, Fe, Zn, W, Zr, In and Ti and the surface of the inorganic oxide fine particle is modified by an organic silane compound represented by the following formula (1) to have hydrophobic groups R^1 and R^2 on the surface:



wherein R^1 is an alkyl group which may be substituted by a methacryloxy group or glycidyloxy group, R^2 is an alkyl group, X is a hydrolyzable group, a is 0, 1 or 2, and b is 1, 2 or 3, with the proviso that $a + b = 3$.

The above oxide may be an oxide of a single element or a composite oxide of two or more elements. Examples of the

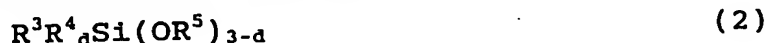
inorganic oxide include silicon oxide, aluminum oxide, tin oxide, antimony oxide, ~~thallium~~ ^{tantalum} oxide, cerium oxide, lanthanum oxide, iron oxide, zinc oxide, tungsten oxide, zirconium oxide, indium oxide, titanium oxide, composite oxide of titanium oxide/zirconium oxide and composite oxide of titanium oxide/zirconium oxide/silicon oxide. The composite oxide preferably contains at least 50 wt% of titanium oxide. The inorganic oxide preferably has a particle diameter of 1 to 100 nm.

10 Examples of the organic silane compound represented by the above formula (1) include methyl trimethoxysilane, γ -acryloxypropyltrimethoxysilane, γ -acryloxypropyltriethoxysilane, γ -acryloxypropylmethyldimethoxysilane, 15 γ -acryloxypropylmethyldiethoxysilane, γ -methacryloxypropyltrimethoxysilane, γ -methacryloxypropyltriethoxysilane, γ -methacryloxypropylmethyldimethoxysilane, γ -methacryloxypropylmethyldiethoxysilane, 20 trimethylchlorosilane, glycidyloxymethyltrimethoxysilane, α -glycidyloxyethyltrimethoxysilane, γ -glycidyloxypropyltrimethoxysilane and γ -glycidyloxypropylmethyldiethoxysilane.

25 The modification of the above oxide fine particle by the organic silane compound may be carried out by hydrolyzing the organic silane compound in the presence of the inorganic oxide particle and a catalyst in a lower alcohol such as ethanol under the reflux of the lower alcohol. As for the ratio of the oxide fine particle to the organic silane compound, the 30 organic silane compound is used in an amount of preferably 0.4 to 2.5 parts by weight, more preferably 0.5 to 1.5 parts by weight based on 1 part by weight of the inorganic oxide.

The inorganic oxide fine particle of the sol (B) is a SiO_2 fine particle whose surface is modified by a hydroxy

film of the above liquid primer composition. The hard coat is a silicon resin-based film, for example, a film of a composition containing (D) a sol of an inorganic oxide fine particle selected from the group consisting of oxides of Si, Al, Sn, Sb, Ta, Ce, La, Fe, Zn, W, Zr, In and Ti and having a particle diameter of 1 to 100 nm and (E) an epoxy-containing silicon compound represented by the following formula (2) or a hydrolyzate thereof:

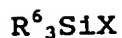


wherein R^3 is a group having an epoxy group and 2 to 12 carbon atoms, R^4 is an alkyl group or haloalkyl group having 1 to 6 carbon atoms, alkenyl group having 2 to 6 carbon atoms, phenyl group or halophenyl group, R^5 is a hydrogen atom, alkyl group having 1 to 4 carbon atoms or acyl group, and d is 0, 1 or 2.

The above sol of an inorganic oxide improves the hardness, heat resistance and weatherability of the hard coat. The inorganic oxide fine particle containing an oxide of Ti or Zr increases the refractive index of the hard coat to a value close to the refractive index of the above primer coat to prevent the generation of an interference fringe. Examples of the above inorganic oxide include SiO_2 , Al_2O_3 , SnO_2 , Sb_2O_5 , Ta_2O_5 , CeO_2 , La_2O_3 , Fe_2O_3 , ZnO , WO_3 , ZrO_2 , In_2O_3 and TiO_2 . These inorganic oxides are preferably in the form of a fine particle having a particle diameter of 1 to 100 μm .

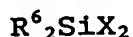
These inorganic oxides are surface treated with an organic silane compound as required to enhance dispersibility in a solvent. The surface treatment may be carried out with an organic silane compound or a hydrolyzate thereof. The organic silane compound is preferably used in an amount of 20 wt% or less based on the inorganic oxide.

Examples of the organic silicon compound include compounds represented by the following formulas (3), (4), (5) and (6).



(3)

wherein a plurality of R^6 's may be the same or different and each an organic group having an alkyl group, phenyl group, vinyl group, methacryloxy group, mercapto group, amino group or epoxy group, and X is a hydrolyzable group.



(4)

wherein R^6 and X are as defined in the above formula (3), with the proviso that a plurality of X's may be the same or different.



(5)

wherein R^6 and X are as defined in the above formula (3).



(6)

wherein X is as defined in the above formula (3).

Examples of the compound represented by the above formula (3) include trimethylmethoxysilane, triethylmethoxysilane, trimethylethoxysilane, triethylethoxysilane, triphenylmethoxysilane, diphenylmethoxysilane, phenyldimethylmethoxysilane, phenyldimethylethoxysilane, vinylldimethylmethoxysilane, vinylldimethylethoxysilane, γ -acryloxypropyldimethylmethoxysilane, γ -methacryloxypropyldimethylmethoxysilane, γ -mercaptopropyldimethylmethoxysilane, γ -mercaptopropyldimethylethoxysilane, N- β (aminoethyl) γ -aminopropyldimethylmethoxysilane, γ -aminopropyldimethylmethoxysilane, γ -aminopropyldimethylethoxysilane, γ -glycidoxypropyldimethylmethoxysilane, γ -glycidoxypropyldimethoxyethoxysilane and β -(3,4-epoxycyclohexyl)ethyldimethylmethoxysilane.

Examples of the compound represented by the above formula (4) include dimethyldimethoxysilane, diethyldimethoxysilane, dimethyldiethoxysilane, diethyldiethoxysilane, diphenyldimethoxysilane, phenylmethyldimethoxysilane, phenylmethyldiethoxysilane, vinylmethyldimethoxysilane, vinylmethyldiethoxysilane,

CLAIMS

1. A transparent laminate comprising a transparent plastic substrate and a coating film formed on at least one side of the substrate, wherein

the coating film is formed by applying and curing a liquid primer composition comprising:

(A) a self-emulsifiable emulsion of a linear polyurethane having a pendant carboxylic acid group and no crosslinked structure between polymer chains;

(B) a sol of an inorganic oxide having a hydrophobic group on the surface; and

(C) a compound having 5 to 9 carbon atoms and at least one hydroxyl group and at least one oxygen atom other than an oxygen atom constituting a hydroxyl group in the molecule.

2. The transparent laminate of claim 1, wherein a coating film of the self-emulsifiable emulsion (A) has an elongation of 240 to 500 % and a 100 % modulus of 190 kgf/cm² or more,